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ATTEMPT TO DEMONSTRATE THE EXISTENCE OF CHEMICALLY BOUND NEUTRONS

by

Sherman Fried, Arnold Friedman,
and Morris Wahlgren

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ABSTRACT

T. J. Grant and J. W. Cobble have reported the delayed emission of neutrons from a cooled (4°K) crystal of LiF which contained trapped electrons and which had been exposed to the neutron beam from a Cockcroft-Walton generator. When the irradiated crystal was removed from the liquid-helium cooling bath and allowed to warm up in a neutron counter, approximately two neutrons could be observed per experiment. These neutrons were emitted at a rate corresponding to a half-time in LiF of approximately 30 sec. These experiments suggested that the neutrons from a Cockcroft-Walton generator were somehow bound in the LiF crystal at low temperatures and were released as the crystal became warm. Attempts were made to duplicate these experiments using conditions as nearly identical as possible, even to the extent of using a LiF crystal supplied by Grant and Cobble. No effect such as they describe could be observed.

INTRODUCTION

In a 1969 paper, Grant and Cobble[1] reported the observation that neutrons could apparently be stored in cooled crystals of LiF that contained large numbers of F centers. Other investigators[2-4] have reported unsuccessful attempts to duplicate these experiments. However, where Grant and Cobble used moderator 14-MeV neutrons from a Cockcroft-Walton generator, other experimenters used moderated thermal neutrons from a reactor or from a cyclotron reaction. On the assumption that differences in the moderated neutron spectra could account for the reported discrepancy in results, we attempted to duplicate as closely as possible the experiment of Grant and Cobble.

During our experiments, we were in communication with Professor Cobble, and his cooperation was greatly appreciated.

EXPERIMENTAL PROCEDURE

The experiments can be divided into two sets.

Set I

In the first set of experiments, the moderator used was a 23-in.-diam paraffin cylinder with a 5-in. well in the center. A quartz dewar flask, placed in the well, was the actual container for the LiF crystal during runs. The configuration is shown in Fig. 1.

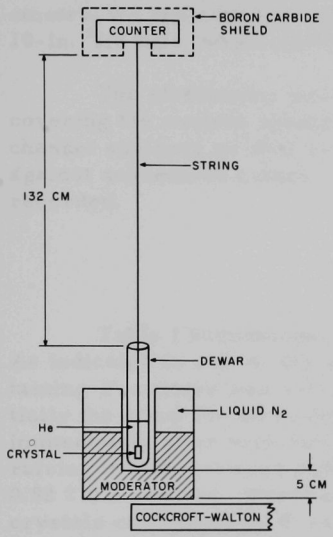


Fig. 1. Diagram of Experimental Arrangement

When the moderator was placed in position as shown, it was cooled to liquid-nitrogen temperatures as measured by a thermocouple imbedded in the paraffin. The flux, as measured by the activation of a gold wire, assuming all the neutrons were moderated to 77°K, varied between 1 and 1.3 x 10^6 n cm^{-2} sec^{-1}. The detectors in these experiments were two high-pressure ^{3}He-filled proportional counters, approximately 1 x 10 cm, arranged parallel to each other in such a way that the exposed crystal could be drawn between them. This gave a counting geometry of about 3%. This was the same counting system used by Krohn et al.[2]

During an irradiation, the quartz dewar was filled with liquid helium and then placed in the well in the cooled moderator, which was partially filled and maintained with liquid nitrogen. The crystal, on a string, was precooled in the liquid nitrogen before being immersed in the liquid helium of the quartz dewar. At the end of a period of radiation, the Cockcroft-Walton was turned off and the string was pulled, drawing the crystal rapidly from the liquid helium into contact with the detectors 5 ft away. This could easily be accomplished within 5 sec. Since the moderator and the detectors were inside the Cockcroft-Walton cave, while the electronics and controls were outside, this arrangement enabled us to start counting much faster than the 30-sec interval required by Grant and Cobble and other experimenters. The LiF crystals in these experiments were both normal lithium fluoride and essentially pure ^{7}LiF. The gamma irradiations used to make the F centers in these crystals were carried out using a ^{60}Co source or the Argonne Linac.

Set II

Professor Cobble was kind enough to lend us a normal LiF crystal, which had yielded "bound neutrons" in his laboratory. This crystal was about 1 x 3 x 0.1 cm and was wrapped with plastic tape. For these experiments, a smaller moderator was built which more or less matched the moderator that Cobble used in experiments with this crystal. Gold wires were irradiated in our Cockcroft-Walton accelerators and exchanged so that the fluxes for counting were matched within 5%. To increase the geometry, we replaced the high-pressure ^{3}He tubes by a low-pressure 3 x 10-in. ^{3}He tube, which resulted in a calculated geometry of 11%.

The electronics were improved so that each pulse in the energy band covering the neutron spectrum was recorded simultaneously on a multichannel analyzer as well as on a scaler. This allowed better discrimination against nonneutron pulses. The time of arrival of each individual pulse was recorded.

RESULTS

Table I summarizes the results of the experiments in the first set. As indicated in row A, the average of 19 experiments with all crystals containing F centers was 1.05 ± 0.3 counts per run, this average being essentially the same for all crystals used. Row B shows the backgrounds obtained immediately after each run by cooling the crystal in liquid helium and raising it to the counter without exposure to neutrons. The average count is 0.82 ± 0.2 per run. However, 10 neutron irradiations were made using LiF crystals containing no F centers. The result, as indicated in Row C, was 0.2 ± 0.15 count/run. Backgrounds obtained in the same manner as above on these crystals are indicated in Row D and were 0.4 ± 0.2 count/run.

TABLE I. Summary of Irradiation Results

Material Irradiated	Flux, n cm^{-2} sec^{-1}	Neutrons Observed, count/run
A. Crystals containing f centers	1.3×10^6	1.05 ± 0.3
B. Crystals containing f centers	No neutrons	0.82 ± 0.2
C. No f centers	1.3×10^6	0.2 ± 0.15
D. No f centers	No neutrons	0.4 ± 0.2

When the output of the amplifier was examined with an oscilloscope, one-third to one-half of the pulses occurring from crystals containing F centers were of abnormal shape and oscillatory rather than single pulses. These spurious pulses may have been caused by sonic vibrations due to relief of strains while warming. Before proceeding with the experiments of Set II, we modified the electronics to reject these pulses.

Table II summarizes the results obtained in the second set of experiments. Within statistical uncertainty, there is no difference between the results obtained with the LiF crystal and the blank. The delayed-neutron events in the blank apparently originate in the surrounding environment, not necessarily the moderator. These events decayed with about a 10-sec half-life. Figure 2 indicates the decay rates of both the crystal and background events.

TABLE II. Effect of Delay in Commencement of Counting

Material Irradiated	Counting Rate, count/sec
A. Crystal irradiated; counted 5-15 sec after irradiation	0.16 ± 0.05
B. Crystal irradiated; counted 15-90 sec after irradiation	0.013 ± 0.005
C. Crystal irradiated; counted 30-90 sec after irradiation	0.007 ± 0.004
Background	
D. Moderator alone irradiated; counted 5-15 sec after irradiation	0.025 ± 0.05
E. Moderator alone irradiated; counted 15-90 sec after irradiation	0.017 ± 0.005
F. Moderator alone irradiated; counted 30-90 sec after irradiation	0.007 ± 0.004

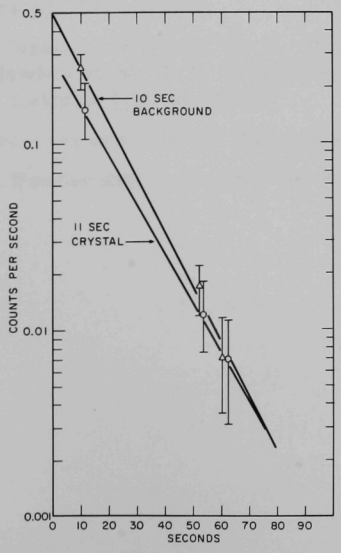
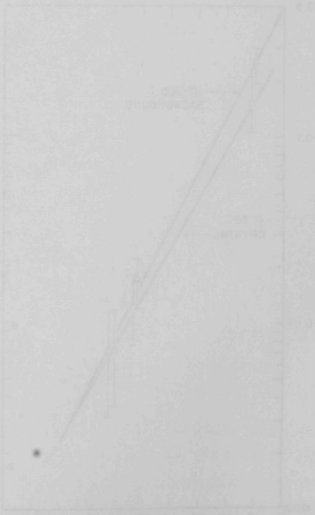


Fig. 2

Decay of Neutron Emission



DISCUSSION

In neither set of experiments could a significant difference be found between background runs and those with crystals. In the experiments described for Set I, the excess over background (measured by crystals without F centers) is apparently due to less refined electronics. In Set II, although a short-lived neutron decay was seen, there is no significant difference between the background and the crystals.

The only remaining difference between our work and Grant and Cobble's is the fact that they used a LiI scintillation detector instead of a ^{3}He detector. It is therefore possible that the differences in the observations between them and ourselves is due to the counting equipment.

ACKNOWLEDGMENTS

We wish to express our thanks to Messrs. L. R. Rawson and A. C. Youngs for the many hours spent in redesigning the Cockcroft-Walton accelerator.

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